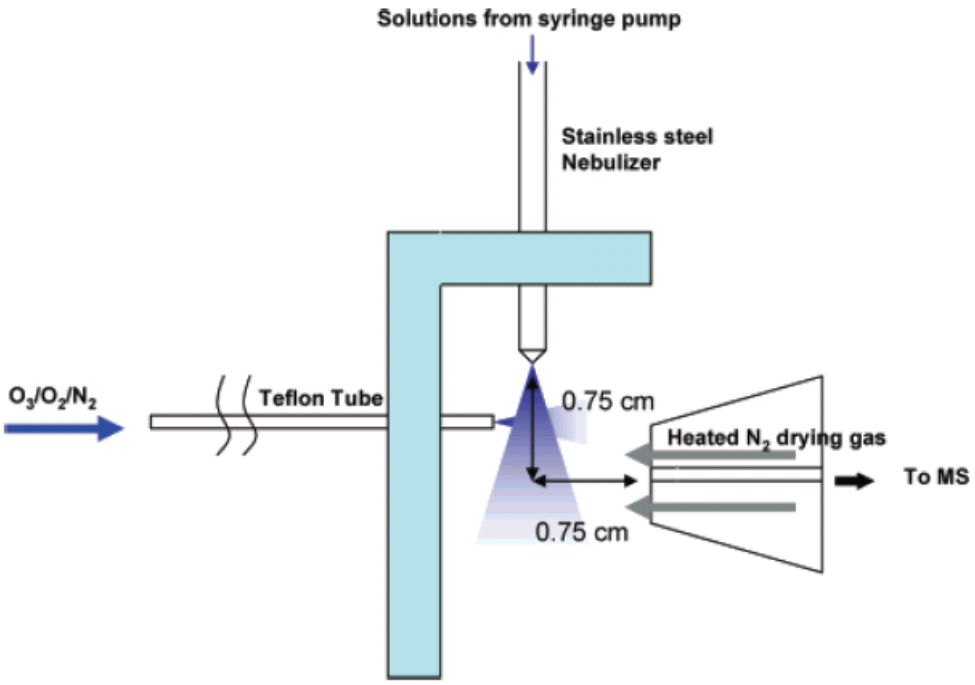
Supporting Information: Droplets produced by fragmentation of electrically neutral liquids, as in the present case, are charged, nevertheless, due to statistical fluctuations that scale with (drop size)$^{-1/2}$. Although the ensemble of spray droplets is on average neutral, individual droplets carry charges that adhere to a Gaussian distribution centered about zero charge, as expected for the random fragmentation of a neutral fluid.[1] This largely unrecognized phenomenon is, however, the basis of Millikan's classical oil drop experiment on the magnitude of the elementary charge,[2] but it is not circumscribed to the laboratory. Thus, for example, the finer sea water aerosol droplets, i.e., those that remain airborne long after being generated during bubble bursting, are knowt to be, on average, negatively charged.[3] This global phenomenon induces a net electrical current that into the atmosphere.[4,5] It should be emphasized that spontaneous, asymmetrical charge separation during pneumatic nebulization of liquids does not generate highly charged droplets, such as those produced in the electrospray ionization of droplets issuing from high-field nozzles.[6,7]

Sprayed droplets eventually undergo solvent evaporation, a process regulated by ambient temperature and relative humidity that increases electrostatic repulsion among excess surface charges. At some point, the repulsion energy exceeds the surface energy of the droplets, whereby they become mechanically unstable and undergo Coulomb explosions in which interfacial charge and mass are shed into smaller droplets.[8-10]These events, when replicated by the progeny droplets, ultimately generate smaller particles that are multiplicatively enriched, i.e., enrichment $\propto (f_i)^m$, where m is the number of successive Coulomb explosions, in the tensioactive species.

Figure S1. Schematic diagram of the spraying chamber, O_3 (g) injection, and mass spectrometer sampling inlet.



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